

Design of recycling system for poly(methyl methacrylate) (PMMA). Part 1: recycling scenario analysis

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Abstract

Introduction In this series of papers, we present a poly(methyl methacrylate) (PMMA) recycling system design based on environmental impacts, chemical hazards, and resource availability. We evaluated the recycling system by life cycle assessment, environment, health, and safety method, and material flow analysis.

Purpose Previous recycling systems have not focused on highly functional plastics such as PMMA, partly because of lower available volumes of waste PMMA compared with other commodity plastics such as polyethylene or polypropylene. However, with the popularization of PMMA-containing products such as liquid crystal displays, the use of PMMA is increasing and this will result in an increase in waste PMMA in the future. The design and testing of recycling systems and technologies for treating waste PMMA is therefore a high research priority. In this study, we analyze recycling of PMMA monomers under a range of scenarios.

Methods Based on the differences between PMMA grades and their life cycles, we developed a life cycle model and designed a range of scenarios for PMMA recycling. We obtained monomer recycling process inventory data based on the operational results of a pilot plant. Using this process inventory data, we quantified life cycle greenhouse gas (LC-GHG) emissions and fossil resource consumption, and we calculated the LIME single index.

Results and discussion PMMA produces more than twice the amount of GHG emissions than other commodity resins. Through scenario and sensitivity analyses, we demonstrated that monomer recycling is more effective than mechanical recycling. Operational modifications in the monomer recycling process can potentially decrease LC-GHG emissions.

Conclusions Highly functional plastics should be recycled while maintaining their key functions, such as the high transparency of PMMA. Monomer recycling has the potential to achieve a closed-loop recycling of PMMA.

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Abbreviations

BHET	Bis(hydroxyethyl) terephthalate
CR	Chemical recycling
DMT	Dimethyl terephthalate
HEA	Home electrical appliances
H-sheet	High molecular weight PMMA sheet
LIME	Japanese life-cycle impact assessment method based on endpoint modeling
LGP	Light guide panel
L-pellet	Low molecular weight PMMA pellet
L-sheet	Low molecular weight PMMA sheet
MMA	Methyl methacrylate
MR	Material recycling (mechanical recycling)
OAE	Office automation equipment
PE	Polyethylene
PET	Polyethylene terephthalate

PMMA	Poly(methyl methacrylate)
PP	Polypropylene
PS	Polystyrene
PTA	Purified terephthalic acid
RPF	Refused plastic/paper fuel
TR	Thermal recycling (thermal recovery)

1 Introduction

Recycling of plastics is an important issue. Examples of previous work in this area include life cycle assessment (LCA) of an Italian plastic recycling system as solid waste (Arena et al. 2003a, b), waste management of soft drink packaging systems in Mexico (Romero-Hernández et al. 2009), and the transboundary recycling of polyethylene terephthalate (PET) bottles between Japan and China (Nakatani et al. 2010). Clift (1997) categorized plastic recycling technologies into reuse, mechanical recycling, depolymerization or monomer recycling, chemical recycling and pyrolysis, and energy recovery. Many alternative processes have been reported and assessed such as recycling options for the management of plastic packaging wastes (Perugini et al. 2005) and feedstock recycling in blast and electric furnaces as materials for reducing oxidized iron to replace cokes (Shimada et al. 2005). The quality of products derived from waste plastics recycled materials was analyzed for evaluating substitutability of fresh plastics made from fossil resources (Nakatani et al. 2011; Nakatani and Hirao 2011) and developing a robust recycling system (Fujii et al. 2012). In this regard, however, the plastics treated in these studies are mainly commodity resins such as polyethylene (PE), polypropylene (PP), polystyrene (PS), polyvinyl chloride (PVC), and PET, which are the most commonly used plastics (JPIF 2012; Plastics 2011).

In addition to high-volume plastics such as PE and PP, other plastics in use have special purposes. Poly(methyl methacrylate) (PMMA) is one such specially used plastic because of its significantly high transparency, resistance to weather and impact, and technical properties. The shipment of PMMA makes up about 1–2 % of the total shipments of all plastics in Japan (JPIF 2012) and Europe (Plastics 2011), and PMMA is used in products requiring high-level transparency and resistance, e.g., automobile lamp covers, lighting equipment, optical fibers, and light guide panels (LGP) of liquid crystal displays (LCDs). With the increased popularity of LCD products, it is likely that the use and stored amount of PMMA has also increased. Although the amount of waste PMMA included in such appliances will also increase in the future, there are currently no effective recycling systems for PMMA. Doddiba et al. (2008) studied the recycling of plastic waste from discarded television sets and Hischier and Baudin (2010) conducted LCA of plasma televisions; however, no previous analysis of the plastics used in LGP is available. The PMMA

parts of electronic devices were not included in the previous analyses such as the recycling in Japan of household appliances and other consumer electronics by Nakano et al. (2007) and Andrae and Andersen (2010).

No reports are currently available on the end of life of PMMA contained in LCD products. It seems that PMMA waste in Japan may be mixed with other plastic wastes such as shredder residue and then treated in the same ways as other commodity resins. Mixed plastics may not be recycled effectively and the quality of recycled materials needs careful assessment to increase recycling efficiency in plastic recycling systems as studied by Nakatani and Hirao (2011). In this regard, however, recycled materials by mechanical recycling of PMMA mixed with other plastics may not make full use of the features of PMMA such as its high transparency. Because of the enforcement of recycling laws and regulations for home electronic devices (METI 2001) and recycling laws for personal computers (PC3R Promotion Association, Japan 2003), LCD display panels contained in such electronic devices are now being collected and are available for recycling.

Monomer recycling of plastics is a technology with the potential to enable closed-loop recycling. Some monomer recycling technologies have been reported and assessed, e.g., monomer recycling of PET bottles in Japan (Sugiyama et al. 2006). For PET bottles, monomer recycling plants were constructed for bottle-to-bottle closed-loop or bottle-to-fiber open-loop recycling (Nakatani and Hirao 2011; Nakatani et al. 2010; Sinha et al. 2010). For PMMA, Achilias (2007) and Lopez et al. (2010) studied pyrolysis using different reactor types in the laboratory. A pilot plant revealed that collected PMMA waste can be recycled to methyl methacrylate (MMA) monomer and then used for producing new PMMA products (MRC 2011; Nakagawa 2007; Sasaki et al. 2008). Several experiments have been done for characterizing its applicability as actual recycling process (Nakagawa 2007).

In this study, we aim to design a PMMA recycling system with a PMMA monomer recycling plant. In practice, several assessments are required for appropriate implementation of new technology into practice, including three types of evaluation of global environmental impacts, local chemical risks, and feasibility studies (Kikuchi and Hirao 2009). In this series of papers, we assess the acceptability of a PMMA recycling process as a social technology by an evaluation of different aspects of the PMMA monomer recycling process. Firstly, we conduct an LCA to quantify global environmental impacts originating from the PMMA life cycle with or without the inclusion of a monomer recycling plant (MRC 2011). Recycling scenarios are set as the different recycling routes in Japan. We consider the substitutability of recycled products for all recycling scenarios. Secondly, we consider operational modifications to improve the performance of the monomer recycling process and the impact on the total life cycle.

2 Materials and methods

2.1 Monomer recycling process

Figure 1 shows the process block flow diagram of PMMA pyrolysis (MRC 2011). The process contains four segments, i.e., depolymerization, liquid recovery (which is the treatment process for effluent gas from the reactor), purification of the MMA monomer, and heat recovery from residue. The raw material for this process is waste PMMA flakes and the product is recycled MMA monomer (99.8 % purity). The utility is heavy oil and electricity, and the inputted amount of which depends on the recovered heat from in-process residue. In the PMMA pyrolysis reactor, PMMA is decomposed by heat into MMA monomer and other substances such as methyl isobutyrate, methyl acrylate, and 1,4-cyclohexane dicarboxylic acid dimethyl ester, i.e., MMA dimer (Kaminsky and Franck 1991; Scheirs and Kaminsky 2006). In the block flow diagram shown in Fig. 1, the liquid recovery segment comprises spray and liquid separation processes. The temperature of effluent gas, named as cracking gases in Fig. 1, is increased from 350 to 500 °C at atmospheric pressure and, after cooling to about room temperature, the crude MMA monomer obtained is condensed before purification of MMA monomer. The purification segment is composed of two distillation columns, where the first column removes low-boiling-point chemicals and the second column purifies the MMA monomer. All residues from the liquid recovery and purification segments are used as fuel in the heat recovery segment for heating sand used as a fluidized bed in the pyrolysis reactor. Although the physical properties of recycled MMA at 99.8 % purity are almost the same as those of fresh MMA, recycled and fresh monomers have been mixed and used for optical devices in actual production. This is because such devices require high transparency enough for graphical function such as color reproducibility, and the use of recycled MMA has a possibility to result in the unpredictable loss or reduction of

the function of optical device by 0.2 % of impurity in recycled MMA. Regarding contamination or impurities in the waste PMMA stream, it was experimentally demonstrated that the MMA monomer can be obtained with the same yield based on the inputted amount of PMMA, even if other types of resins are mixed with waste PMMA (Nakagawa 2007; METI and MRC 2008; Sasaki 2008). Metals and other organic materials, e.g., paper, cause no problems in the reaction (Nakagawa 2007; Ministry of Economy et al. 2008; Sasaki 2008). Impurities are collected as residue or decomposed in the pyrolysis reactor, converted into low molecular weight substances, and incinerated in the heat recovery incinerator. This means that contamination with these types of impurities increases the recovered heat. In this regard, however, PVC may be an inhibitor and an undesirable contaminant of PMMA recycling by pyrolysis (Scheirs and Kaminsky 2006).

2.2 PMMA life cycle model based on plastic grade

Figure 2 shows the life cycle model of PMMA contained in products. PMMA has two main grades as a polymer: high and low molecular weight polymer grades. These two grades of PMMA have slightly different physical properties. Although high and low molecular weight PMMAs are both incorporated in the same type of products (LGP in LCD panel), the size or other features of them are different. In Fig. 2, we have specified three types of PMMA materials based on their categorization in Japanese statistical data (Japan Petrochemical Industry Association JPIA 2012), i.e., high molecular weight sheet (H-sheet), low molecular weight sheet (L-sheet), and low molecular weight pellet (L-pellet). The products made from them are labeled as products A, B, and C, respectively. The sheet-type PMMA provides the raw materials for LGP of LCD products such as television sets, PC monitors, laptops, and mobile phones. Additionally, board products produced from PMMA sheet include components of furniture and information panels. On the

Fig. 1 Outline of a PMMA pyrolysis process pilot plant

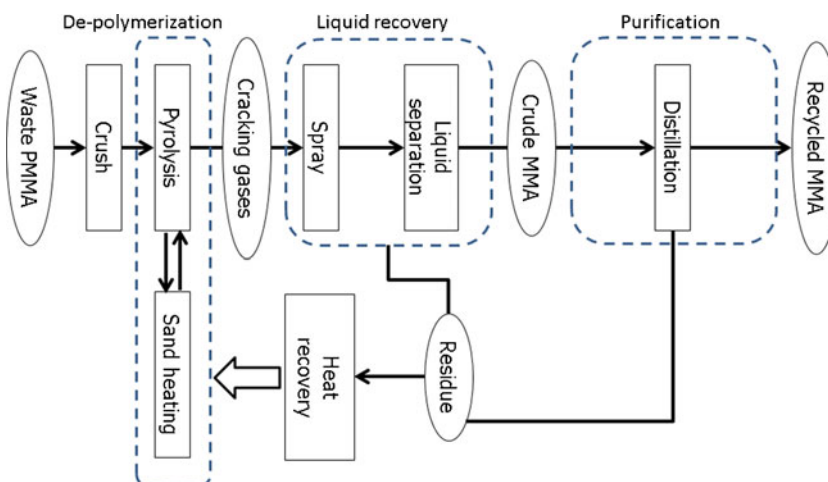
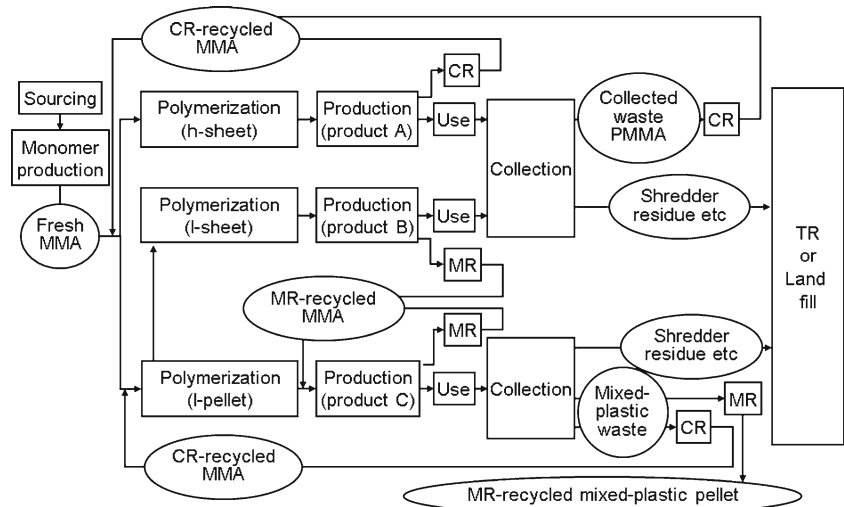


Fig. 2 Life cycle model of two grades of PMMA developed in Japan



other hand, various categories of plastic products incorporate pellet-type PMMA, including machine tools and parts, daily necessities, and miscellaneous goods (Ministry of Economy et al. 2012). This is because the L-pellets can be molded and formed into designed shapes. L-sheet is one of the products from L-pellets. In this regard, however, H-sheet is produced by direct polymerization of the MMA monomer and cannot be produced by molding, because high molecular weight PMMA cannot be melted to form a shape for products.

After PMMA products are used, they must be collected on the individual pathways based on the condition of waste products containing PMMA. Existing recycling laws in Japan enable the collection of products A and B in Fig. 2. Most of the PMMA product contained in LCD panels may be collected and treated in recycling plants (Hirasawa 1999). An investigation of an appliances recycling plant in Japan revealed that LGPs in LCDs were gathered and accumulated as by-products from the manual separation of valuable parts and metals in LCD products (Fig. 3). LGP is almost 100 % pure PMMA after removing dots of titanium oxide attached on the surface of the LGP. PMMA boards included in construction waste can also be collectable by segregating them from other boards. In this regard, however, we define the collection stages of H- and L-sheets as the same process as shown in Fig. 2. Collected waste products A and B are transported to chemical recycling (CR) meaning monomer recycling, thermal recovery (TR), or landfill, but not to mechanical recycling (MR) because the high molecular weight PMMA cannot be recycled mechanically and it is not easy to distinguish between grades of PMMA in collection sites and recycling plants. The TR or landfilling process is the same in Japan as for other plastic recycling, which includes the production and use of refuse paper/plastic fuel (RPF), energy recovery in a cement kiln, waste power generation, incineration without energy recovery, and landfilling (Mayumi et al. 2010; PWMI 2010). On the other hand, molding materials, i.e., product C in Fig. 2, may be

collected not as PMMA but as mixed plastics. According to an investigation of a treatment plant for scrapped cars, specific parts made of a single resin such as bumpers are segregated for recycling to increase the ratio of car recycling. L-pellets, for example, are used in containers and packaging, and it is quite difficult to segregate PMMA products from other materials. PMMA wastes mixed with other types of plastics can be transported to CR, MR, TR, or landfilling processes as shown in Fig. 2. A certain amount of waste PMMA can be recycled to MMA monomer with the same ratio as products A and B in the CR process. R and landfilling are the same processes for product C as for products A and B.

2.3 Evaluation settings

2.3.1 Functional unit and impact category

We defined the functional unit of LCA in this paper as the use of one unit amount of PMMA, i.e., 1 kg in Japan. The composition of products A, B, and C was set at the same ratio as the actual shipment of them based on PMMA statistics



Fig. 3 Waste LGPs made of PMMA piled inside a home appliance recycling plant

(Japan Petrochemical Industry Association JPIA 2012). The ratios have fluctuated slightly over the last 10 years. In this study, we adopted the 2009 ratios as representative values, being 0.173, 0.131, and 0.696 kg for products A, B, and C, respectively. In previous studies, environmental impacts of the plastic life cycle were analyzed based on greenhouse gas (GHG) emissions (Nakatani et al. 2010) with fossil resource consumption (Mayumi et al. 2010) or other impact categories (Shena et al. 2010). In this study, these indicators were taken into account to the assessment of PMMA recycling process. As for the impact categories, we evaluated Japanese life-cycle impact assessment method based on endpoint modeling (LIME) single index with the consideration of ozone depletion, global warming, acidification, air pollution in urban areas, photochemical oxidant creation, emission of hazardous chemicals, ecotoxicity, eutrophication, and solid waste (Andrae 2009; JLCA 2012).

2.3.2 Case study 1: recycling pathways

In this case study, we quantified the recycling effects of PMMA pyrolysis in the life cycle of PMMA. Table 1 displays the recycling scenarios in case study 1. Scenario 1 is set as the reference case, where all waste PMMA is incinerated without energy recovery. In scenario 2, production loss is adequately recycled, i.e., high and low molecular weight polymer grades are recycled in CR and MR, respectively. PMMA waste after use is transported for TR or landfilling, including the recycling effects of RPF, cement raw material/fuel, and waste power generation and the impacts of incineration and landfilling. The distribution ratio for each process is assumed to be the same as the average ratio for industrial plastic waste, and these values were extracted from the statistical data in 2009 (PWMI 2010). In scenario 3, the waste PMMAs from products A and B

are sent to CR. Because the amount of PMMA in the form of product C is larger than those of products A and B, the scenario settings on the waste from product C were generated as scenarios 3 and 4. Under scenario 3, we assume the recycling of collected product C in the MR process. At that time, collected PMMA waste and other plastics are mixed. According to the existing literature and reports (JCPRA 2007), such mixed plastic pellets are used to produce plastic pallets, which can reduce the amount of fresh PP, but not PMMA. Therefore, the recycling effects of mechanically recycling mixed plastic to mixed-plastic pellets can be considered as the substitution of fresh PP. As the process inventory for PMMA mechanical recycling, we adopted the data from mechanical processing of PS (JLCA 2012) because PS sheet is applicable to similar types of molding products, which means that their heat and mechanical properties might be similar. In mechanical recycling of PS, waste PS is crushed, melted, and pelletized to produce recycled PS pellet. In contrast, all PMMA waste is recycled in CR, and MMA monomer is output in scenario 4. Recycled MMA can be substituted for fresh MMA. We obtained the process inventory data of PMMA monomer recycling from the operation results of an actual pilot plant in June of 2009. Other recycling process inventory data were obtained from the Japanese LCA database (JEMAI 2007; JLCA 2012) and the literature on plastic recycling (JCPRA 2007).

We defined other settings for LCA as shown in Table 1. The transportation distance in all scenarios was set as the same value, 200 km, to compare recycling methods without the effect of location. This distance includes all required transportation after PMMA product assembly. We defined the treatment processes for waste PMMA products by using available inventory data. From the existing database (JLCA 2012), we determined two types of treatment processes for office automation equipment (OAE) and home electrical appliances (HEA): mixing

Table 1 Scenario settings in two case studies. Scenarios 1 to 4 are the settings in case study 1 and scenario 5 shows the settings of case study 2

Scenario			1	2	3	4	5
Plastic recycling settings	Production loss	H-MW	Incineration	CR (yield, 70 %)	CR (yield, 70 %)	CR (yield, 70 %)	CR (yield, 77 %)
		L-MW	Incineration	MR	MR	MR	MR
	After use of products A and B		Incineration	RPF/ cement raw material and fuel/ waste power generation/ incineration/ landfilling	CR (yield, 70 %)	CR (yield, 70 %)	CR (yield, 77 %)
		After use of product C	Incineration		MR (mixed-plastic pellet)		
Other settings	Transportation distance (km)		200				
	Waste treatment process	Products A and B	Average of mixing treatment processes for OAE and HEA			Average of advanced treatment processes for OAE and HEA	
		Product C	Average of mixing treatment processes for OAE and HEA and dismantling process for automobiles			Average of advanced treatment processes for OAE and HEA and dismantling process for automobiles	

and advanced treatment processes. OAE and HEA treatment processes include disassembling, scraping, shredding, and sorting processes, where valuable parts, metals, and plastics are separated. In the mixing treatment, plastics contained in OAE and HEA are outputted as shredder dust, whereas they are outputted as separated plastic wastes from advanced treatment process. A part of PMMA contained in product C is an automobile component and the inventory data of treatment processes for scrapped cars are available (JLCA 2012). In this study, the inventory data for treating unit amounts of waste products A, B, and C were assumed as the averages of OAE, HEA, and cars as shown in Table 1. All required background inventory data were extracted from the Japanese LCA database (JEMAI 2007; JLCA 2012) and the literature on plastic recycling (JCPRA 2007).

2.3.3 Case study 2: operational modification of the PMMA monomer recycling process

The waste residue from the monomer recycling process is utilized as fuel on site as shown in Fig. 1. This approach may reduce fuel consumption and GHG emission. The waste residue from the purification segment in Fig. 1 includes high amounts of MMA, which is also incinerated for heat recovery. PMMA contains oxygen in its molecular structure, which leads to less heat generation than from PE or PP, whereas PMMA has higher cumulative environmental loads per unit amount of production than the other resins (PWMI 2009). The advantage of monomer recycling to the thermal recycling is relatively higher than other plastics in case of PMMA. If the yield of MMA to the unit amount of waste PMMA is increased by operational modification, energy available for the recycling process is reduced because of the decrease in residue used for fuel. In this study, we modified the purification segment comprising two distillation columns to specify the

optimum purification settings and minimize the environmental impacts. We compared the effect of the increased recovery of recycled MMA with the increase in fuel consumption.

Chemical process simulators can be employed to estimate the inventory of chemical process units (Iosif et al. 2010). Aspen Plus V7.3 (Aspen Technology, Inc., Burlington, MA, USA), a widely utilized process simulator used in previous LCA studies (Iosif et al. 2010; Mendivil et al. 2006), was selected in this study for estimating inventories of modified processes. The two columns for purifying the MMA monomer were simulated to estimate process inventory data. Other processes such as the pyrolysis reactor, spray, and flasher were not changed and their inventory data are set as the actual pilot plant data. Figure 4 shows the simulation model developed on Aspen Plus. The boundary of simulation was set as the purification and heat recovery units in Fig. 1. Crude MMA in Fig. 4 includes components with lower and higher boiling points than pure MMA; the boiling point of which is about 101 °C. To separate such components, two distillation columns are utilized, where lower and higher boiling components are separated from MMA in first and second columns, respectively. The RadFrac rigorous distillation model was selected and applied as the model for simulation in the Aspen Plus. The incineration process for treating separated lower and higher boiling components, i.e., flows 2 and 5 in Fig. 4 from columns, and heat recovery was also simulated by an incinerator model in Aspen Plus. A pre-heater is inserted before feeding mixed residue to incinerator for increasing temperature sufficiently. Additionally, heat integration through a whole processes was conducted based on pinch analysis (Biegler et al. 1997). The minimum temperature between hot and cold streams was over 100 °C, which means that the recovered fuel is sufficiently available to meet the heat duty in this purification part. To compensate for the degreased residue, i.e., fuel oil additive for pyrolysis part, by increasing MMA yield, additional

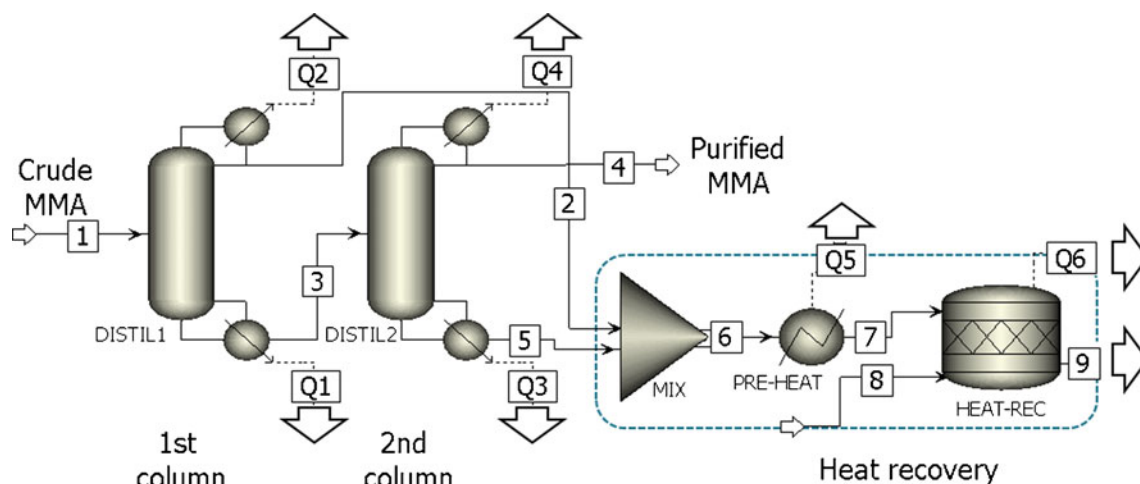


Fig. 4 Simulation model of MMA purification process developed on Aspen Plus

fuel oil was utilized. The limitation of specification of devices and instruments, such as limit temperature of the material used for heat exchanger, was defined based on process engineering heuristics (Trambouze 2000). The number of stages and feeding stage were fixed to the same settings as the actual pilot plant. The MMA yield was controlled by changing the distillate rate of the second column, and the reflux ratios of both columns and bottom rate of the first column were set as the optimization parameters to minimize total energy demand. The simulation was performed with the change in recovery yield of MMA monomer to waste PMMA from 0.7, value at the actual pilot plant, to 0.77, the highest value achievable by distillation (see also “Discussion” section). At that time, simulation results did not correspond with the actual plant results because of the simplification of actual energy demand, which includes energy demands for air conditioners, illumination, and cascade recycling of steam on site. To bridge such gaps,

the coefficient of simulation result and actual plant at the same operation and plant settings was calculated and applied to estimate the process inventory from simulation results at different settings. The LCA settings were organized in Table 1 as scenario 5, where the yield was set as the highest value to check the process with the largest recovery of MMA.

3 Results

Scenarios 1 to 4 in Fig. 5a show the results of case study 1. Scenario 1 shows the total life cycle greenhouse gas (LC-GHG) per functional unit without recycling methods. It has the highest values, which means that all recycling processes can reduce LC-GHG emission. The reduction in the impact shown in scenario 2 demonstrates the recycling effects of thermal recovery from waste PMMA. LC-GHG emissions

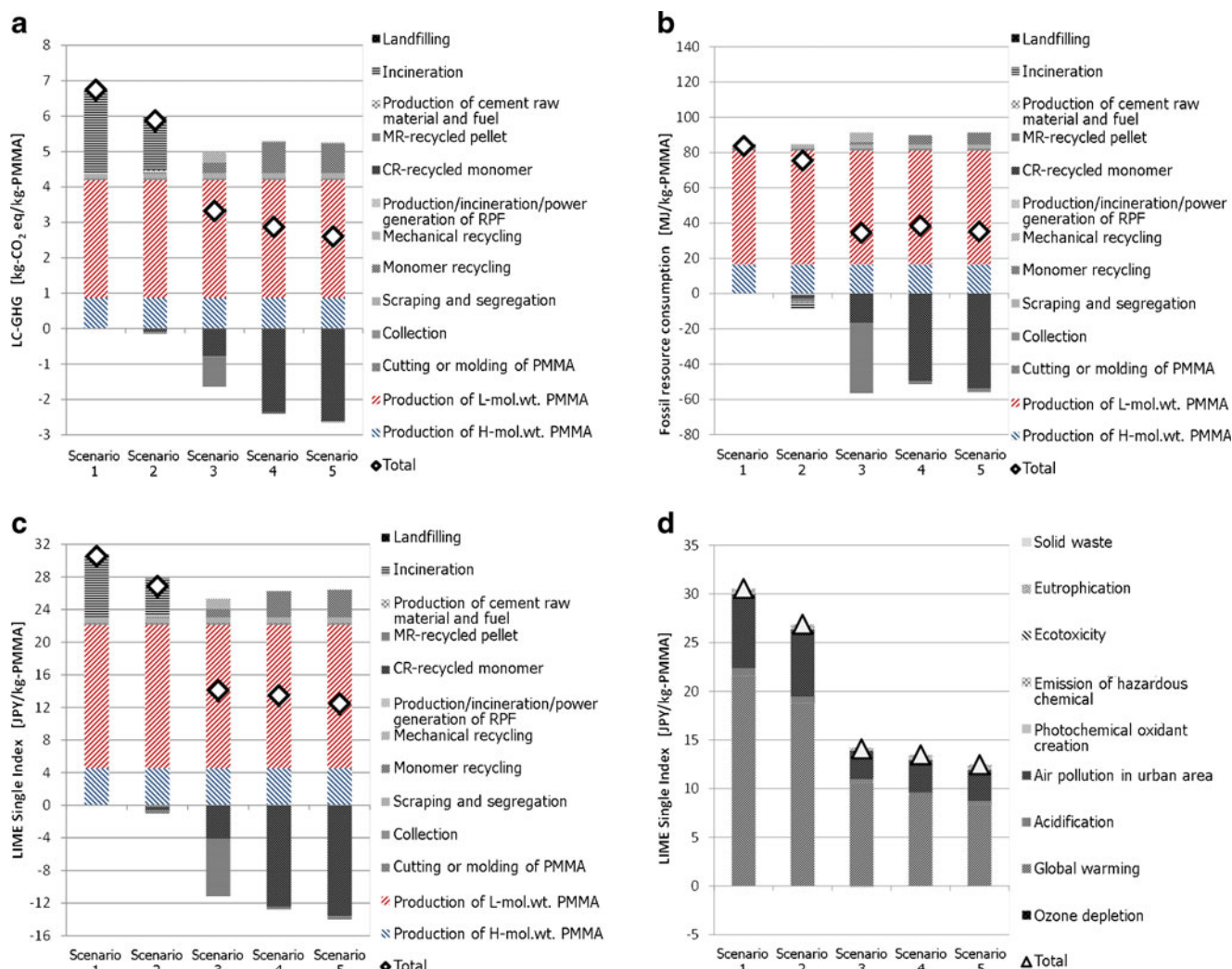


Fig. 5 Assessment results of scenarios in case studies 1 and 2. **a** LC-GHG. **b** Fossil resource use. **c** LIME single index (life cycle stage). **d** LIME single index (impact category)

were reduced by about 10 % in scenario 1 by using recovered heat as a substitute for fossil fuel. The impact of PMMA recycling on production loss is observed in the results of scenario 2, where about 4 % of LC-GHG in scenario 1 is mitigated. Because scenario 4 has lower LC-GHG emissions than scenario 3, PMMA monomer recycling has a higher ability to reduce LC-GHG emissions than does mechanical recycling. The substitution of PMMA or PP leads to the difference in the recycling effects of monomer recycling and mechanical recycling, even though monomer recycling requires more resources than does mechanical recycling. Hence, the PMMA monomer recycling process has the highest priority in all recycling processes in terms of LC-GHG emissions. Scenarios 1 to 4 in Fig. 5b–d show the results of case study 1 as assessed by fossil resource consumption and the LIME single index. The recycling effects of MR and CR are different in terms of GHG emissions and fossil resource consumption as shown in the results of scenarios 3 and 4. It leads to the different order of scenarios 3 and 4 in GHG emission and fossil resource consumption. As for the LIME single index, the ranking of scenarios shows the same tendency as found in the LC-GHG result, whereas the gap between scenarios 3 and 4 is slightly decreased in the result indicated by the LIME single index. Global warming potential has the largest contribution to the LIME single index, and air pollution in urban areas is the second largest impact category.

Figure 5 shows the results of scenario 5, case study 2, where operational modifications were tested. It demonstrates that the increased yield of MMA monomer from 0.7 to 0.77 results in a decrease in environmental impacts, fossil resource consumption, and the LIME single index. This means that the recycling effect of increased recovery of MMA in monomer recycling is higher than the negative effect caused by the requirement for increased fuel. Because of the high environmental load of production of PMMA from fossil fuels, the increased yield of MMA monomer in the PMMA pyrolysis process leads to an increase in the effect of monomer recycling on GHG emission, fossil resource use, and the LIME single index, even if additional fuel energy is required for recovering MMA monomer from crude MMA in distillation columns.

4 Discussion

The environmental loads and fossil resource consumption for the production of PMMA and PP are different. Although GHG emissions from PMMA production are more than twice as high as emissions from PP production, other environmental loads, especially SO_x , NO_x , and fossil resource consumption, are not significantly different. These differences in environmental loads and GHG emissions are reflected in the different trends in the results indicated by LC-GHG, fossil resource

consumption, and the LIME single index. When analyzing recycling effects, the yields of recycled materials in PMMA recycling plants have a comparatively large effect on the final results, which are set as 0.853 for the yield of PS mechanical recycling (JLCA 2012) and 0.7 for mechanical and monomer recycling processes, respectively. The sensitivity of a 10 % change in the yield of mechanical recycling, i.e., 0.768 to 0.938, to LC-GHG, fossil resource consumption, and the LIME single index was 2.65, 11.8, and 5.06 %, respectively. In this regard, the fossil resource consumption in scenario 3 becomes almost the same as that in scenario 4 when the yield of mechanical recycling is set at 0.768. This means that if the yield from mechanical recycling is lower than 0.768, the priority of scenarios 3 and 4, based on consumption of fossil resources, is reversed. Because some mechanical recycling processes have much lower yields, e.g., 0.521, in a process for waste containers and packaging (JLCA 2012), monomer recycling of PMMA may also be a priority in terms of fossil resource consumption.

Existing systems of the collection and segregation of waste plastics are applicable for some types of product containing PMMA. As mentioned in the “Materials and methods” section, LCDs made from PMMA are collected and piled in the HEA recycling process. The recycling process (Hirasawa 1999) is regulated by the Home Appliance Recycling Act in Japan (METI 2001) and it has a role in the collection of valuable materials such as rare earth elements (Yamasue et al. 2009). Such systems can be adapted for the collection of PMMA. For other molding products made from l-PMMA pellets, existing recycling systems such as cars, construction materials, and containers and packaging can also be adapted to collect PMMA. PMMA pyrolysis has an advantage in that the yield of MMA to inputted waste PMMA is not greatly changed by contamination with other materials (MRC 2011; Nakagawa 2007; Sasaki et al. 2008); therefore, PMMA pyrolysis is a robust recycling process (Fujii et al. 2012) for the possibility of contaminating various types of plastics in waste. Note that molding products containing PMMA have not been investigated in detail, and so, further investigation and accumulation of statistical data are required.

The results shown in Fig. 5 demonstrate the recycling effect of PMMA pyrolysis. PMMA pyrolysis recycling may be more efficient than mechanical recycling as shown in Fig. 5a, c–d. PMMA tends to be used for products requiring excellent transparency and climate resistance properties, and mechanically recycled PMMA pellets cannot achieve such high performance. With increasing requirements for plastics to replace conventional heavy materials such as glass or metal (Humbert et al. 2009; Ribeiro et al. 2006), PMMA can be used as a substitute for glass. LCD panels will be required to meet the demand for television sets (Display Search 2012). An adequate recycling method for waste PMMA is required, and PMMA pyrolysis can be an effective method for fulfilling the closed-loop recycling of MMA all over the world.

The high effectiveness of PMMA monomer recycling is partly because PMMA is composed of a single monomer. PET, for example, has two monomers: ethylene glycol and terephthalic acid. Although the monomer recycling of PET is also effective, the process flow of PMMA pyrolysis shown in Fig. 1 is simpler than that of PET depolymerization through purified terephthalic acid (PTA) and dimethyl terephthalate (DMT) processes (Sugiyama et al. 2006). The complexity of chemical processes leads to high-energy consumption due to the increase of process units such as reactors and separators, which was shown in PET monomer recycling processes (Sugiyama et al. 2006). Although BHET process in PET monomer recycling has small number of reactor and separator comparing with PTA and DMT processes, the monomer recycling of PET is regarded as it has higher environmental impacts than the mechanical recycling of PET (Nakatani et al. 2010). Monomer recycling of PMMA has an advantage in environmental impacts than mechanical recycling of PMMA.

To improve monomer recycling performance, MMA included in the crude MMA flow should be recovered as much as possible. In case study 2, yields were increased from 0.7 to 0.77 by adopting a change in the operation of the two distillation columns. The largest value in the distillation columns settings in this case study was 0.77 because of the difficulties in purifying MMA monomer by distillation. Considering the amount of MMA contained in the flow from the pyrolysis reactor, the maximum yield can be increased to 0.88. Other process options, such as an increase in the number of distillation column stages or other types of separation, e.g., extraction and adsorption units, can increase the yield of recycled MMA. Therefore, the PMMA monomer recycling process has an opening point for process improvement of environmental impacts. If process changes are made in the future, solvent and adsorbent must be carefully selected with a consideration of their impacts in their life cycles (Nguyen et al. 2009).

5 Conclusions

The development of a specialized recycling process for PMMA can reduce the environmental impacts originating from its life cycle. PMMA monomer recycling is the highest priority for reducing LC-GHG and other environmental impacts. Our investigations of an existing recycling plant for home electronic appliances show that existing processes can be applied for collecting waste PMMA contained in light guide panels without additional process modifications. Other recycling systems for construction materials, automobiles, and containers and packaging have the potential to collect waste PMMA, although segregation processes from other types of resins must be addressed. Additionally, operational modifications in the monomer recycling process to increase the yield of the MMA monomer can reduce total environmental impacts

even further, in spite of the additional fuel consumption used in MMA purification. This is because the production of new MMA has a high environmental impact and recycling MMA is effective in reducing the overall life cycle impact.

Although the total amount of PMMA use is not large in plastics, a closed-loop recycling system, which monomer recycling may achieve, can be established together with recycling of other materials contained in these products such as base or rare metals (Yamasue et al. 2009). This was confirmed in this study through investigations of recycling plants for home electronic appliances and automobiles, where each component of the product is segregated as much as possible by manual procedures (Hirasawa 1999) or by specialized scrapping machines. The PMMA monomer recycling process can contribute to the establishment of a recyclable society.

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